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"Managing phonon transport by core/shell nanowires"

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Abstract:

We study thermal conductivity in core-shell nanowires (NW) by using molecular dynamics simulations. Interestingly, a remarkable oscillation effect in heat current autocorrelation function in core-shell NWs has been observed, while the same effect is absent in pure silicon nanowires, nanotube structures, and random doped nanowires.

Detailed characterizations of the oscillation signal reveal that this intriguing oscillation is caused by the *coherent resonance* effect of the transverse and longitudinal phonon modes. This phonon resonance results in the localization of the longitudinal modes, which leads to the reduction of thermal conductivity in core-shell nanowires.

Our study provides a novel coherent mechanism that can be used to tune thermal conductivity in core-shell nanowires.

Introduction:

Thermoelectric materials with low thermal conductivity are favorable for high-efficiency power generation and refrigeration. In the past few years, some significant progresses have been achieved in enhancing thermoelectric performance of silicon, which is abundant in nature and well engineered in semiconductor industry, and friendly to the environment. For example, by etching the surface of silicon nanowires (SiNWs), it has been demonstrated that thermal conductivity of bulk silicon can be reduced more than two orders of magnitude with slight effect on electric power factor, resulting in a dramatically enhanced figure of merit (ZT) at room temperature.

However, most of the conventional approaches to reduce thermal conductivity, such as introduction of rough surface and impurity scatterings, are based mainly on incoherent mechanisms, which cause phonons to lose coherence, this in turn also deteriorates the electronic transport properties. Recent experimental works have demonstrated that, by altering phonon band structure in periodic nanomesh structures, a remarkable enhancement in ZT can be obtained by

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14. ABSTRACT
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significantly reducing the thermal conductivity of silicon while preserving its electrical conductivity. These studies have offered new perspective to improve *ZT* based on coherent mechanisms.

In our work, we demonstrate an intriguing phonon coherent resonance phenomenon in Ge/Si core-shell NWs, which offers a coherent mechanism to tune thermal conductivity in core shell NWs. As Ge/Si core-shell NWs can be synthesized

Computational Simulation:

The configuration of [100] Ge/Si core-shell NWs studied is shown in Fig. 1. The cross sections of both core and shell regions are square, with side length L_c and L, respectively. The Stillinger–Weber (SW) potential is used in our equilibrium molecular dynamics (EMD) simulations to derive the force term. For Si-Ge bond, the net length and energy units in SW potential are taken to be the arithmetic average and geometric average of that of Si and Ge atom, respectively. The velocity Verlet algorithm is employed to integrate Newton's equations of motion numerically, and time step is set as 0.8 fs. The details techniques can be found in the publication [1].

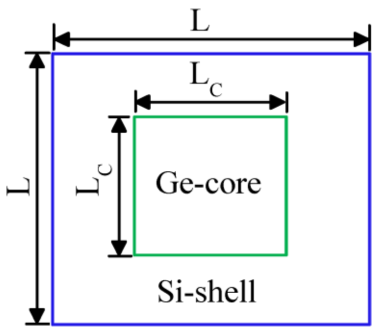

Figure 1. Cross sectional view of [100] Ge/Si core-shell NWs. The cross sections of both core and shell regions are square, with L_c and L denoting the side length of core and shell regions, respectively.

Results and Discussions:

Phonon Coherent Resonance

A novel coherent resonance phenomenon has been observed in core/shell nanowire. It is reflected in the time dependence of normalized heat current autocorrelation function (HCACF) shown in Fig. 2. It is quite obvious that such phenomenon is absent in pure Si nanowire, Si nanotube, and Si/Ge nanowire.

We perform the Fourier Transform analysis of the HCACF and found that the coherent resonance come from the phonon confinement by the core/shell structure, as is demonstrated in Fig 3.

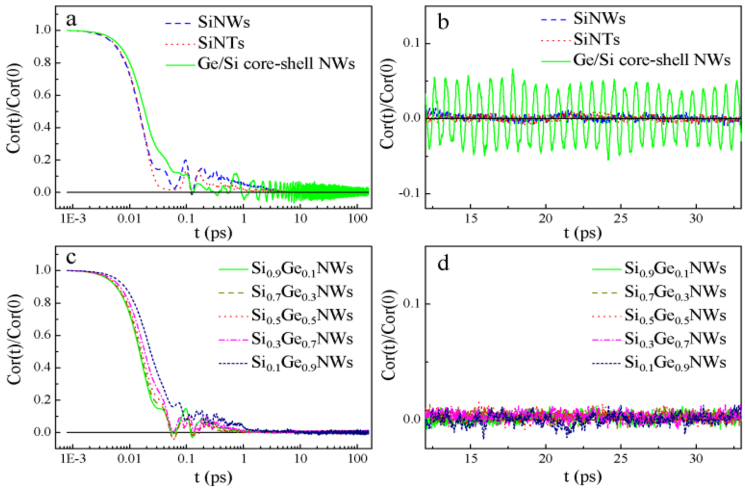

Figure 2. Time dependence of normalized heat current autocorrelation function (HCACF). a, Normalized HCACF for SiNWs (blue dash line), SiNTs (red dot line) and Ge/Si core-shell NWs with L_c/L=0.65 (green solid line). b, Long-time region of a. c, Normalized HCACF for $Si_{1-x}Ge_x$ NWs with different doping concentration x. d, Long-time region of c. The black lines in all figures draw the zero axis for reference. Here the super cell size is 16×5×5 and temperature is 300 K.

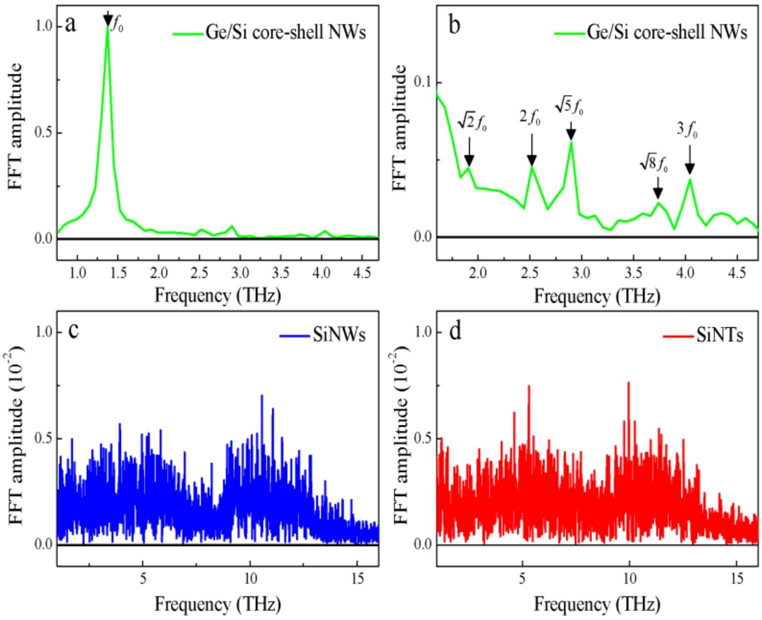

Figure 3. Amplitude of the fast Fourier transform (FFT) of the long-time region of normalized HCACF. The black lines in all figures draw the zero axis for reference. a, Ge/Si core-shell NWs. b, The high frequency oscillation peaks for Ge/Si core-shell NWs. The black arrows pinpoint the different oscillation frequencies. c and d are amplitudes of the FFT of the long-time region of normalized HCACF for SiNWs and SiNTs, respectively.

Turning thermal conductivity by the Core/Shell structure

Due to the nonpropagating nature of the transverse modes, the resonance effect induced by coupling between the transverse and longitudinal modes can hinder the longitudinal transport, thus offering a coherent mechanism to tune thermal conductivity in core-shell structure.

To illustrate this coherent mechanism in more details, we plot in Fig 4 the localization of phonon modes (characterized by participation ratio shown in Fig 4 a), and the thermal conductivity (Fig 4 b) versus the core/shell ration L_c/L. It is clearly seen that the thermal conductivity can be tuned by this ratio. More importantly, a minimum is reached for a value of L_c/L, which means there is an optimal ratio to make the lowest thermal conductivity. This is very important and has potential application in designing thermalelectric materials.

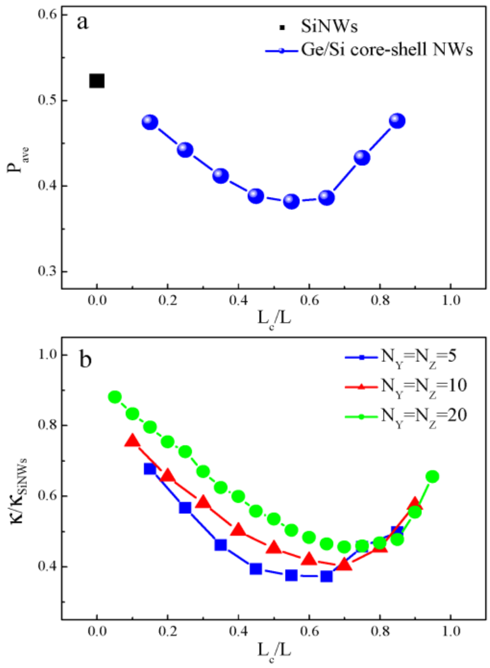

Figure 4. a, Averaged phonon participation ratio P_{ave} versus core-shell ratio in Ge/Si core-shell NWs (blue circle). The black square plots P_{ave} in SiNWs with the same cross section area for comparison. All phonon modes are computed with a cross section of 5×5 unit cells. b, Structure dependence of thermal conductivity in Ge/Si core-shell NWs with different cross section areas ($N_Y \times N_Z$ unit cells) at 300 K. Thermal conductivity of SiNWs with the same cross section area is used as the reference.

In addition to the main work on the core/shell structures nanowires, we have also studied thermal conductivity of silicon nanowires (SiNWs) with different cross sectional geometries [2]. It is found that thermal conductivity decreases monotonically with the increase of surface-to-volume ratio (SVR). More interestingly, a simple universal linear dependence of thermal conductivity on SVR is observed for SiNWs with modest cross sectional area (larger than 20 nm2), regardless of the cross sectional geometry. As a result, among different shaped SiNWs with the same cross sectional area, the one with triangular cross section has the lowest thermal conductivity.

We also studied thermal conduction through the silicon dioxide/silicon interface which is published in [3].

Because of our group's significant contribution to heat conduction in low dimensional nanoscale systems in past years, we have been invited to write a Colloquium by *European Physical Journal B* [4]. The support of AOARD has been acknowledged.

Overall, we have gone beyond what we outlined in the original proposal. The support from AOARD is kindly acknowledged in all my invited talks in many international conferences etc.

List of Publications and Significant Collaborations that resulted from your AOARD supported project:

1. J. Chen, G Zhang, and B Li, Phonon coherent resonance and its effect on thermal transport in core-shell nanowires, *The Journal of Chemical Physics*, **135**, 104508, 14 September 2011.
2. J. Chen, G Zhang, and B Li, A universal gauge for thermal conductivity of silicon nanowires with different cross sectional geometries, *The Journal of Chemical Physics*, **135**, 204705, 29 November 2011.
3. J. Chen, G Zhang and B Li, Thermal contact resistance across nanoscale silicon dioxide and silicon interface, *Journal of Applied Physics*, **112,** 064319, 24 September 2012.
4. S. Liu, X.-F Xu, R. G. Xie, G. Zhang and B. LI, Anomalous heat conduction and anomalous diffusion in low dimensional nanoscale systems, *European Physics Journal B* **85**, 337, 1 October 2012.

Conference Presentation:
The work has been presented in the Thermal Science Grantee's Meeting, 26-28 September 2011, Arlington VA, organized by Dr. Kumar Jata from the US Airforce.

Attachments: Publications 1-3 are attached.

DD882: As a separate document, please complete and sign the inventions disclosure form.

Important Note: If the work has been adequately described in refereed publications, submit an abstract as described above and refer the reader to your above List of Publications for details. If a full report needs to be written, then submission of a final report that is very similar to a full length journal article will be sufficient in most cases. This document may be as long or as short as needed to give a fair account of the work performed during the period of performance. There will be variations depending on the scope of the work. As such, there is no length or formatting constraints for the final report. Keep in mind the amount of funding you received relative to the amount of effort you put into the report. For example, do not submit a $300k report for $50k worth of funding; likewise, do not submit a $50k report for $300k worth of funding. Include as many charts and figures as required to explain the work.